

## **Role of accelerators in the study of heavy ions and superheavies in nature**

**A P Sharma**

Department of Physics, Aligarh Muslim University, Aligarh-202 002, India

**Abstract :** The extremely heavy ions including superheavies (upto charge  $Z \sim 114$ ) are produced in nature (Cosmos/galactic spaces) in nucleosynthesis processes. But the measurement of their charge is extremely complicated. For calibration of track lengths the accelerators play a great role. Various heavy ions upto  $Z=92$  (uranium) produced by the accelerators create track length in meteoritic material (SSNTD) and are used for estimating the unknown charges of the heavy ions irradiating the meteorites while in space for several thousand years. About 20 stony iron meteoritic pallacites (having charge threshold,  $Z \geq 20$ ) have been analysed and partially annealed at high temperatures for several hours in order to create fading in latent tracks and develop a new charge threshold of almost  $Z > 50$  for the study of heavy galactic nuclei and superheavies ( $Z \sim 114$ ) in nature. Scanning of about  $8 \text{ cm}^3$  (volume) of meteoritic crystals gives us about 580 U-group tracks ( $Z=91-95$ ) and 6-7 superheavy elements. This study also infers that the uranium ion concentration in galactic spaces of the cosmos is more as compared to the solar system uranium abundance.

**Keywords :** Accelerators, super-heavy elements, nuclear track detectors, nucleosynthesis.

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### **1. Introduction**

Undoubtedly the studies of extremely heavy ions and superheavies embraced in the meteorites while in space for several thousand years reveal useful information about the processes of nucleosynthesis and energy spectra of the heavy nuclei alongwith the presence of hypothetical superheavy elements (SHE) of charge  $Z \sim 114$  theoretically predicted by Myers and Swiatecki (1966) and Meldner (1967), but the comparison of track lengths formed in space with the track length formed due to accelerated heavy ions (upto charge  $Z=92$ , presently available accelerated heavy ions) is most essential. The nuclei with  $Z \geq 20$  were studied initially in 1967 by Fowler *et al* (1967) using a nuclear emulsion stack with a surface area of  $1 \text{ m}^2$  flown for 24 hours in the upper layers of the atmosphere. These experiments for the search of superheavies were continued by them upto 1977 in stacks of thick layers of nuclear emulsions of large size. Around 1975-77 similar

studies have been performed by the groups of Fleischer *et al* (1975) and Fowler *et al* (1977) with nuclear emulsion and plastic detectors. In this experiment stacks with surface area of  $1.5 \text{ m}^2$  were exposed for 8 months on Sky Lab. Many exposures during a period of 10 years were taken by this group. In these experiments only 23 tracks due to actinide series were observed and not even a single track was found by Fowler *et al* (1977) and Shirk and Price (1978) which could be attributed to an ion of charge,  $Z \geq 110$ . The use of meteoritic crystals detectors was made for the first time by Maurette *et al* (1975). The tracks of stopping galactic nuclei of  $Z \geq 20$  are automatically recorded for about  $10^7$ - $10^8$  years in silicate minerals of various meteorites while in space and reveal information about the galactic heavy ions and the superheavy elements (SHE).

By etching suitable minerals like olivine one can obtain visible tracks whose length parameter depends on the charge of the ion and the nature of the silicate mineral (detector). We have surveyed about 20 stony iron meteorites, but the crystals of Marjalahti, Liporsky, Khutor and Eagle Station meteorites were selected for special investigations of extremely heavy ions and superheavy elements. The olivine crystals from within the unexposed central part of the meteorites were used for heavy ion track length calibration by exposing them to accelerated heavy ion beams of Dubna (USSR) and German cyclotrons. In the present study about  $8 \text{ cm}^3$  olivine crystals from various meteoritic pallasites are studied using the partial annealing method. The length spectrum of volume tracks is measured and the abundances of different nuclei groups are calculated from volume track length spectrum using the L-Z (length-charge) identification method and the calibration curve. The results gave about 580 uranium group ( $Z=91-95$ ) tracks and 6-7 superheavy elements (SHE) tracks besides the lower charge group. The relative abundance of ultraheavy/very very heavy (VVH) cosmic ray nuclei as compared to Fe-group nuclei has also been calculated.

## 2. Experimental and results

The selected crystals of meteorites were mounted in epoxy (Polmer's optimum conditions being  $60^\circ\text{C}$  for 1 hour) and then grinded and polished. Modified  $\text{WO}_4$  solution ( $\text{pH}=7.85$ ) developed by Krishnaswami *et al* (1971) has been used for etching. The olivine crystals are etched at a temperature of  $100^\circ\text{C}$  for 20-30 hours in a hermetically closed volume, while pyroxene crystals are etched in  $\text{NaOH}$  solution (6 gm  $\text{NaOH}$  in 6 cc  $\text{H}_2\text{O}$ ) in teflon containers at  $140^\circ\text{C}$  (boiling point) for few hours (Lal *et al* 1968). The tracks developed after proper etching have been measured for their densities and lengths under optical microscope. The tracks of length  $\leq 20 \mu\text{m}$  are grouped as VH tracks pertaining to Fe-group nuclei. The lengths  $\geq 30 \mu\text{m}$  are counted for VVH track density measurements, although their

abundance is very small ( $VH/VH \sim 10^{-8}-10^{-4}$ ). The measurement of track densities were made for different samples and different locations of the olivine and pyroxene crystals of different meteorites using TINT and TINCLE method (Lal *et al* 1968, Lal 1969 and Sharma *et al* 1981, 1983). The results of Fe-group track density and peak track length spectrum for different meteorites and moon samples are given in Table 1.

**Table 1.** Iron group track density and peak track length measurements in different meteorites and moon samples.

Sl. No.	Nature of samples	Fe-Group track density (t/cm <sup>2</sup> )	Fe-ion group peak track length	
			(i) For fossil Fe-tracks	(ii) For accelerated Fe-ion tracks
<u>Meteorites :</u>				
1.	Tugalin Bulean meteorite	225 × 10 <sup>4</sup>	10-12 μm	18-19 μm
2.	Patwar meteorite	100 × 10 <sup>4</sup>	13-14 μm	18-19 μm
3.	Marjalahti meteorite	102 × 10 <sup>4</sup>	6-8 μm	12-14 μm
4.	Dhajala meteorite	3 × 10 <sup>4</sup>	8-9 μm	13-14 μm
<u>Moon Samples :</u>				
1.	Luna-16	45 × 10 <sup>4</sup>	91-0 μm	14-15 μm
2.	Luna-24	42 × 10 <sup>4</sup>	8-10 μm	14-15 μm

Table 1 compares the fossil Fe-group track length peak with the accelerated Fe-ion track length peaks of monoenergetic energy from cyclotrons at JINR, Dubna, Moscow (USSR) in meteoritic crystals. The accelerated heavy ions ranging from Fe to U of few tens-hundreds MeV energies have been used for these studies. The former fossil Fe-track lengths seem to be shortened in all meteorites as compared to the accelerated Fe-track lengths. This difference can be attributed to some sort of annealing of latent tracks in space. The different amount of shortening in case of different meteorites can be explained due to their being in different orbits in space with respect to the position of the sun.

The identification of ultraheavy (VH) and superheavy element (SHE) tracks has been done using the method of partial annealing. The early study of ultra-heavy cosmic rays using the fossil tracks in meteorite minerals has shown irregular shifting and broadening of the different peaks in track length spectrum, which is attributed to the space fading of fossil tracks. Hence the controlled annealing in the laboratory was considered the only possible way to eliminate this effect (Sharma *et al* 1981, 1983, Yadav *et al* 1983). This method can easily eliminate the background due to Fe-group (VH tracks) and other tracks upto  $Z \sim 50$  and hence allows to perform such studies in relatively small sized crystals.

Table 2 indicates the optimum annealing conditions being maintained in order to eliminate VH track background completely and also to calibrate track length data for charge assignment.

**Table 2.** The optimum annealing condition for various limiting charges.

Sl. No	Annealing condition	Limiting charges annealed
(i)	380°C for 72 hours	$Z < 30$
(ii)	400°C for 60 hours	$Z < 40$
(iii)	410°C for 50 hours	$Z < 45$
(iv)	430°C for 32 hours	$Z < 50$

The volume tracks have been measured under the above conditions and their length reduction is depicted in Figure 1 and also in Table 3.

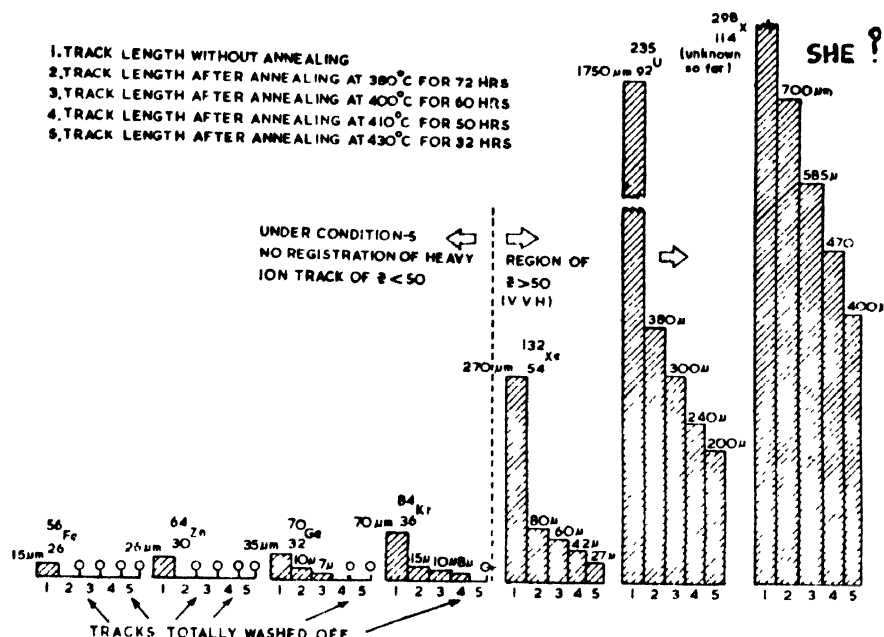
**Table 3.** Track length reduction in meteoritic samples under different annealing conditions for different heavy ion exposures.

Accelerated heavy ion beam (~10 MeV/N to 100 MeV/N)	Exposure from cyclotron at	heavy ion volume track length spectrum with the annealing condition				
		(i) without annealing	(ii) 380°C 72 Hrs	(iii) 400°C 50 hrs	(iv) 410°C 50 hrs	(v) 430°C 32 hrs
$^{56}_{26}\text{Fe}$	JINR, Dubna (USSR)	15 $\mu\text{m}$	Nil	Nil	Nil	Nil
$^{64}_{30}\text{Zn}$	—do—	26 $\mu\text{m}$	Nil	Nil	Nil	Nil
$^{70}_{32}\text{Ge}$	—do—	35 $\mu\text{m}$	10 $\mu\text{m}$	7 $\mu\text{m}$	Nil	Nil
$^{84}_{36}\text{Kr}$	—do—	70 $\mu\text{m}$	15 $\mu\text{m}$	10 $\mu\text{m}$	5 $\mu\text{m}$	Nil
$^{136}_{54}\text{Xe}$	—do—	270 $\mu\text{m}$	80 $\mu\text{m}$	60 $\mu\text{m}$	42 $\mu\text{m}$	27 $\mu\text{m}$
$^{238}_{92}\text{U}$	Darmstadt (Germany)	1750 $\mu\text{m}$	380 $\mu\text{m}$	300 $\mu\text{m}$	240 $\mu\text{m}$	200 $\mu\text{m}$
$^{294}_{114}(\text{SHE})$	Exposure source of SHE is Natural (Cosmos) Cyclotron of the Galactic spaces		700 $\mu\text{m}$	585 $\mu\text{m}$	470 $\mu\text{m}$	400 $\mu\text{m}$

(Unknown/hypothetical element so far)

**Note:** The charge assignment of SHE is done after calibration with known accelerated beams of many ions from  $^{56}_{26}\text{Fe}$  upto  $^{238}_{92}\text{U}$  along with extrapolation beyond U-ion range point.

The curve marked (a) in Figure 2 shows the plot of etchable track length as a function of atomic number (Z) on the basis of the approach of Fieni *et al* (1976) for track identification. The points on this curve represent the experimental track length data of accelerated Fe, Zn, Ge, Kr and U ions (presently available heavy ions of suitable energy producing volume tracks in olivine crystals). The extrapolation



**Figure 1.** Partial annealing of tracks in meteoritic crystals. Only  $^{132}\text{Xe}$ ,  $^{235}\text{U}$  and unknown SHE (superheavy element) tracks can be revealed after annealing at  $\sim 430^\circ\text{C}$  for 32 hours.

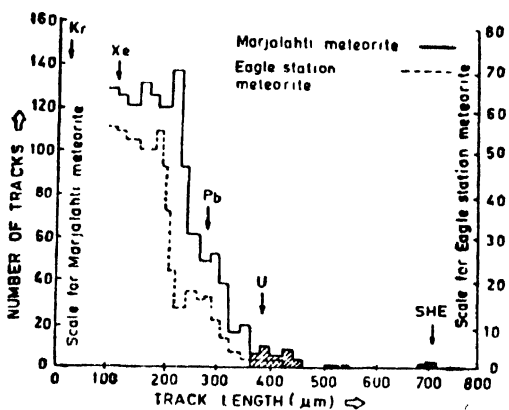
to higher atomic number ( $Z$ ) has been done on the basis of Katz and Kobetich model (1968) of track formation. The curve (b) in Figure 2 shows the variation of track length with  $Z$  under space conditions for faded fossil tracks in olivine crystals, while the curves ( $C_1$ ,  $C_2$ ,  $C_3$  and  $C_4$ ) in Figure 2 shows the variation of track length under different annealing conditions.

Figure 3 shows the Fe-group track length spectra for lunar crystals (Luna-16 and Luna-24). In this case also the peak falls at  $\sim 9.5 \mu\text{m}$ . This also confirms that the fossil tracks in olivine crystals are shortened in comparison to the fresh Fe-tracks due to heavy ion beam from cyclotron. This may be attributed to the high moon temperature ( $\sim 120^\circ\text{C}$ ) during day time, which might have caused some annealing.

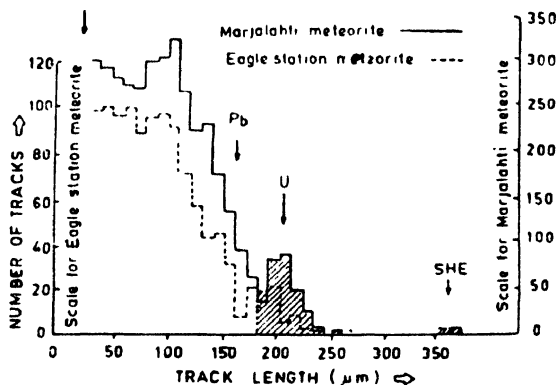
Figures 4 and 5 represent the track length spectra for Marjalahti and Eagle Station meteorite olivine crystals under annealing conditions of  $380^\circ\text{C}$  for 72 hours and  $430^\circ\text{C}$  for 32 hours which show the existence of very long tracks of length  $\sim 700 \mu\text{m}$  and  $350 \mu\text{m}$  respectively. According to our charge assignment, all these long tracks belong to the nuclei with atomic number ( $Z$ )  $\sim 110$ -115 and seem to be the tracks of superheavy elements (SHE).

The results of experiments on the superheavy elements (SHE) synthesis using  $^{48}\text{Ga}$  and  $^{238}\text{U}$  beams are mentioned in the following Table 4.





**Figure 4.** The track length distribution in two different meteoritic crystals under same annealing conditions of 380°C for 72 hrs.



**Figure 5.** The track length distribution in two different meteoritic crystals under same annealing conditions of 430 °C for 32 hrs.

The Table 5 shows the computed magnitudes of the abundances of different charge groups relative to Fe-group.

The uranium group abundance ( $Z=91-95$ ) of our present work (column A) is higher than the solar system abundances (column B). In fact the uranium group nuclei are found more by a factor of about 10 ( $\sim 9.8 \times 10^{-7} / 1.0 \times 10^{-7}$ ) as compared to this group in the solar system abundances. This is in confirmation with the results of Lexan detector experiments by Shirk *et al* (1973) and Price *et al* (1975), which show that the U concentration in galactic spaces is about 10 times of the U-concentration in solar system.

**3. Conclusion**

Our present experimental results claim the existence of superheavy elements (SHE) in nature (using partial annealing method), but some more experimental work

is needed for confirming this new finding. The work also predicts that the Fe-track lengths are shortened in all meteorites as compared to the accelerated Fe-track

**Table 4.** Results of superheavy synthesis using  $^{48}\text{Ca}$  and  $^{238}\text{U}$  cyclotrons beams.

Reaction	Compound nucleus of composite system	Measured life time	Cross section limit ( $\text{Cm}^2$ )	References
<b>With Ca :</b>				
$^{243}\text{Am} + ^{48}\text{Ca}$	$^{291}\text{115}$	2h-1yr	$2 \times 10^{-35}$	Oganessian et al (1978)
$^{242}\text{Pu} + ^{48}\text{Ca}$	$^{290}\text{114}$	2h-1yr	$10^{-35}$	—do—
$^{246}\text{Cm} + ^{48}\text{Ca}$	$^{294}\text{116}$	2h-1yr	$2 \times 10^{-35}$	—do—
$^{248}\text{Cm} + ^{48}\text{Ca}$	$^{296}\text{116}$	2h-1yr	$2 \times 10^{-35}$	—do—
		6h-1yr	$2 \times 10^{-35}$	Otto et al (1978)
		1s-1yr	$8 \times 10^{-35}$	Illige et al (1978)
		$10^{-5}\text{s-5d}$	$1.5 \times 10^{-34}$	Armbruster (1983)
$^{232}\text{Th} + ^{48}\text{Ca}$	$^{280}\text{110}$	$\geq 3\text{ ms}$	$3 \times 10^{-35}$	Ter-Akopian et al (1979)
$^{231}\text{Pa} + ^{48}\text{Ca}$	$^{279}\text{111}$	$\geq 3\text{ ms}$	$4 \times 10^{-35}$	—do—
<b>With U :</b>				
$^{238}\text{U} + ^{238}\text{U}$	$^{476}\text{184}$	1ms-0.5yr	$2 \times 10^{-32}$	Freisleben et al (1979)
		6hr-1yr	$7 \times 10^{-36}$	Gaggeler et al (1980)
$^{248}\text{Cm} + ^{238}\text{U}$	$^{486}\text{188}$	6hr-1hr	$1.5 \times 10^{-34}$	Herrmann (1981)

lengths. Also the uranium content is found more enriched in galactic cosmic spaces in comparison to the solar system abundance (Sharma 1982, 1988).

**Table 5.** Abundances of different charge groups with respect to the Fe-group.

Charge group (Z between)	Abundances calculated from our measurements (A)	Solar system abundances (B)
56-60	$2.8 \times 10^{-6}$	$3.2 \times 10^{-6}$
61-65	$6.2 \times 10^{-6}$	$3.0 \times 10^{-6}$
66-70	$6.0 \times 10^{-6}$	$3.1 \times 10^{-6}$
71-75	$7.2 \times 10^{-6}$	$4.5 \times 10^{-6}$
76-80	$4.5 \times 10^{-6}$	$7.2 \times 10^{-6}$
81-85	$4.6 \times 10^{-6}$	$6.0 \times 10^{-6}$
86-90	$7.5 \times 10^{-6}$	$3.2 \times 10^{-6}$
91-95 (U-group)	$9.8 \times 10^{-7}$	$1.0 \times 10^{-7}$



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